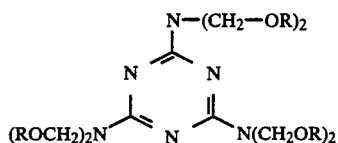


# ALKYLCARBAMYL METHYLATED AMINO-TRIAZINE CROSSLINKING AGENTS AND CURABLE COMPOSITIONS CONTAINING THE SAME

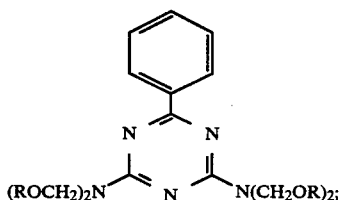
The present invention relates to curing agents, to curable compositions and to methods of making and using them. More particularly, the present invention relates to novel curing agents comprising alkylcarbamylnitraminotriazines and to curable compositions comprising an active hydrogen-containing material, a novel alkylcarbamylnitraminotriazine and a cure catalyst. Coatings cured from the compositions have exceptional resistance to detergent and salt spray exposure, making them well adapted for use in powder coatings, coil coatings and can coatings. The new compositions can be used with filler to provide shaped articles of manufacture with superior properties.

## BACKGROUND OF THE INVENTION

Curable compositions containing aminotriazine compounds are known in the art. As is shown in Koral et al., U.S. Pat. No. 3,661,819, for example, a preferred family of aminotriazine curing agents comprises (i) a triaminotriazine compound of the formula:



which will be depicted hereinafter as  $\text{C}_3\text{N}_6(\text{CH}_2\text{OR})_6$ ; or (ii) a benzoguanamine compound of the formula:



which will be depicted hereinafter as  $\text{C}_3\text{H}_5(\text{C}_6\text{H}_5)(\text{CH}_2\text{OR})_4$  wherein R is hydrogen or alkyl of from 1 to 12 carbon atoms. It is also known to use oligomers of such compounds, which are low molecular weight condensation products containing for example two, three or four triazine rings, joined by  $-\text{CH}_2\text{OCH}_2-$  linkages, as well as mixtures of any of the foregoing. These are used to cure active hydrogen-containing materials, especially polymers which contain carboxyl groups, alcoholic hydroxy groups, amide groups and groups convertible to such groups, such as methylol groups. When such curable compositions are applied to substrates as coatings and then cured, excellent hardness, impact resistance, light stability and solvent resistance is imparted to the articles. The compositions can also be formulated with fillers and/or reinforcements such as particulate and fibrous mineral and organic materials, such as cellulose, wood, glass, graphite, textiles, silica, asbestos, wollastonite, and the like to produce insulation, foundry molds and the like which have superior properties and show a reduced tendency to emit formaldehyde during use.

As is described in German Patent OL No. 2,005,693 (1971) (Chemical Abstracts 76:P 34864 a (1972)), when triaminotriazines of the general formula (i) above are reacted with arylurethanes, such as phenyl urethane, there are produced reaction products of the typical formula  $\text{C}_3\text{N}_6(\text{CH}_2-\text{NH}-\text{COOC}_6\text{H}_5)_6$ , and when these are reacted with polymers containing hydroxyl groups such as acrylics and polyesters, crosslinking occurs with the development of colorless, very hard films, which remain colorless even when the baking time is increased tenfold, to five hours at  $100^\circ\text{C}$ . However, subsequent experiments have shown that such coatings, like those crosslinked with the triazines of formulae (i) and (ii) above, are somewhat deficient in detergent resistance, salt spray resistance and adhesion. They also are produced with the liberation of phenol, which causes health and disposal problems, and is economically wasteful.

It has now been discovered that if aminotriazines of general formulae (i) and (ii) are reacted with alkylurethanes (which are well known to be less reactive than the aryl carbamates used in the above-mentioned German Patent), derivatives are formed which are also reactive to crosslink active hydrogen-containing polymers, but the new coatings which are formed have much improved properties (detergent, salt spray, adhesion, color retention) over those of the prior art, particularly the aryl-substituted derivatives of OL No. 2,005,693.

Although it is known, e.g., from Amin et al., Indian J. Chem., 14B, 139-140 (1976), to prepare both aryl and alkyl carbamylnitramethylated melamines, by the reaction of trimethylolmelamine with n-hexyl carbamate, only the mono-substituted product was produced, and this would not be capable of acting as a crosslinker to introduce two or more urethane groups. Such groups are now believed to be essential to secure all of the advantages of the present invention.

Thus the present invention differs from the state of the art by providing aminotriazine derivatives containing at least two alkylcarbamylnitramethyl groups and then using them as crosslinkers for active hydrogen-containing materials to provide coatings with exceptional resistance, for example, to detergent and salt-spray exposure, and improved light stability.

## SUMMARY OF THE INVENTION

According to the present invention there are provided triazine compounds selected from:

- (i) a triaminotriazine compound of the formula  $\text{C}_3\text{N}_6(\text{CH}_2\text{OR})_{6-x}(\text{CH}_2\text{NHCOOR}^1)_x$ ;
- (ii) a benzoguanamine compound of the formula  $\text{C}_3\text{N}_5(\text{C}_6\text{H}_5)(\text{CH}_2\text{OR})_{4-y}(\text{CH}_2\text{NHCOOR}^1)_y$ ;
- (iii) an oligomer of (i) or of (ii); or
- (iv) a mixture of at least two of any of (i), (ii) and (iii), wherein the R groups are, independently, hydrogen or alkyl of from 1 to 12 carbon atoms, the  $\text{R}^1$  groups are, independently, alkyl of from 1 to 20 carbon atoms, x is in the range of from about 2 to about 6, and y is in the range of from about 2 to about 4.

In preferred embodiments of the invention, x is in the range of from about 2.8 to about 6 and y is in the range of from about 2.2 to about 4. With respect to compound (i) R is lower alkyl, preferably  $\text{C}_1-\text{C}_8$  and  $\text{R}^1$  is methyl, ethyl, n-propyl, i-propyl, butyl, n-octyl, 2-ethylhexyl, n-octadecyl, or a mixture of any of the foregoing. Also preferred are oligomers of (iii)(i) in which R is methyl